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[54] **CRYOGENIC RECTIFICATION SYSTEM FOR PRODUCING HIGHER PURITY HELIUM**

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[52] **U.S. Cl.** **62/639**; 62/923

[58] **Field of Search** 62/639, 923

[56] **References Cited**

U.S. PATENT DOCUMENTS

3,807,185	4/1974	Forg et al.	62/639
4,238,211	12/1980	Stuart	62/639
4,666,481	5/1987	Olson, Jr.	62/11
4,701,200	10/1987	Fisher et al.	62/27
4,701,201	10/1987	Hanson et al.	62/27
5,329,775	7/1994	Fiedler et al.	62/24
5,339,641	8/1994	Mathis et al.	62/639

OTHER PUBLICATIONS

Rathmann et al., *Linde Reports on Science and Technology No. 34, 1982.*

Pahade et al., *Thermal Integrated of Processes to Recover Helium,* Energy Process, vol. 8, No. 4, Dec., 1988 pp. 196–200.

Handley et al., *Process Requirements and Enhanced Economics of Helium Recovery from Natural Gas,* Soc. of Petroleum Engineers SPE 24292, Apr. 13, 1992 pp. 11–20.

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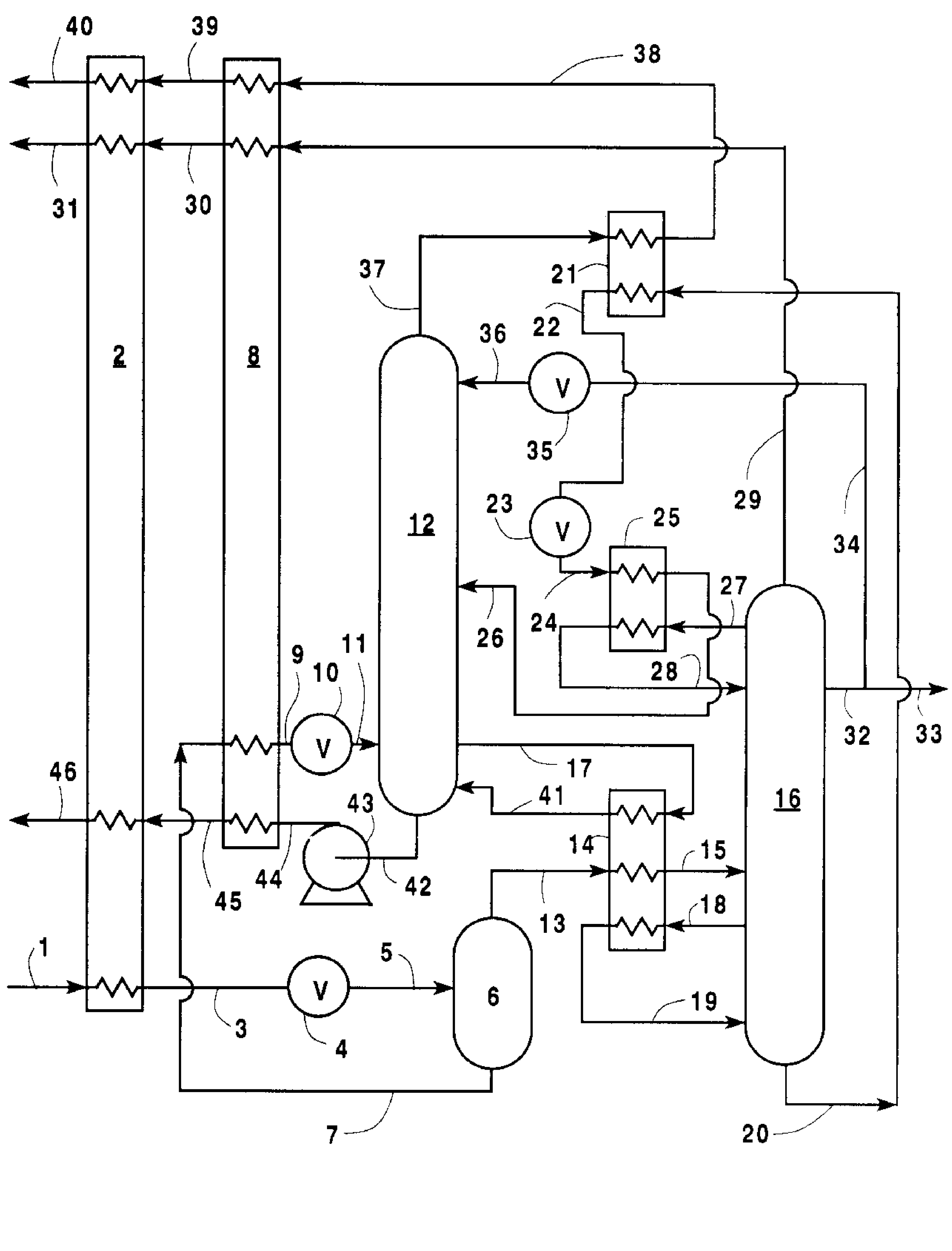
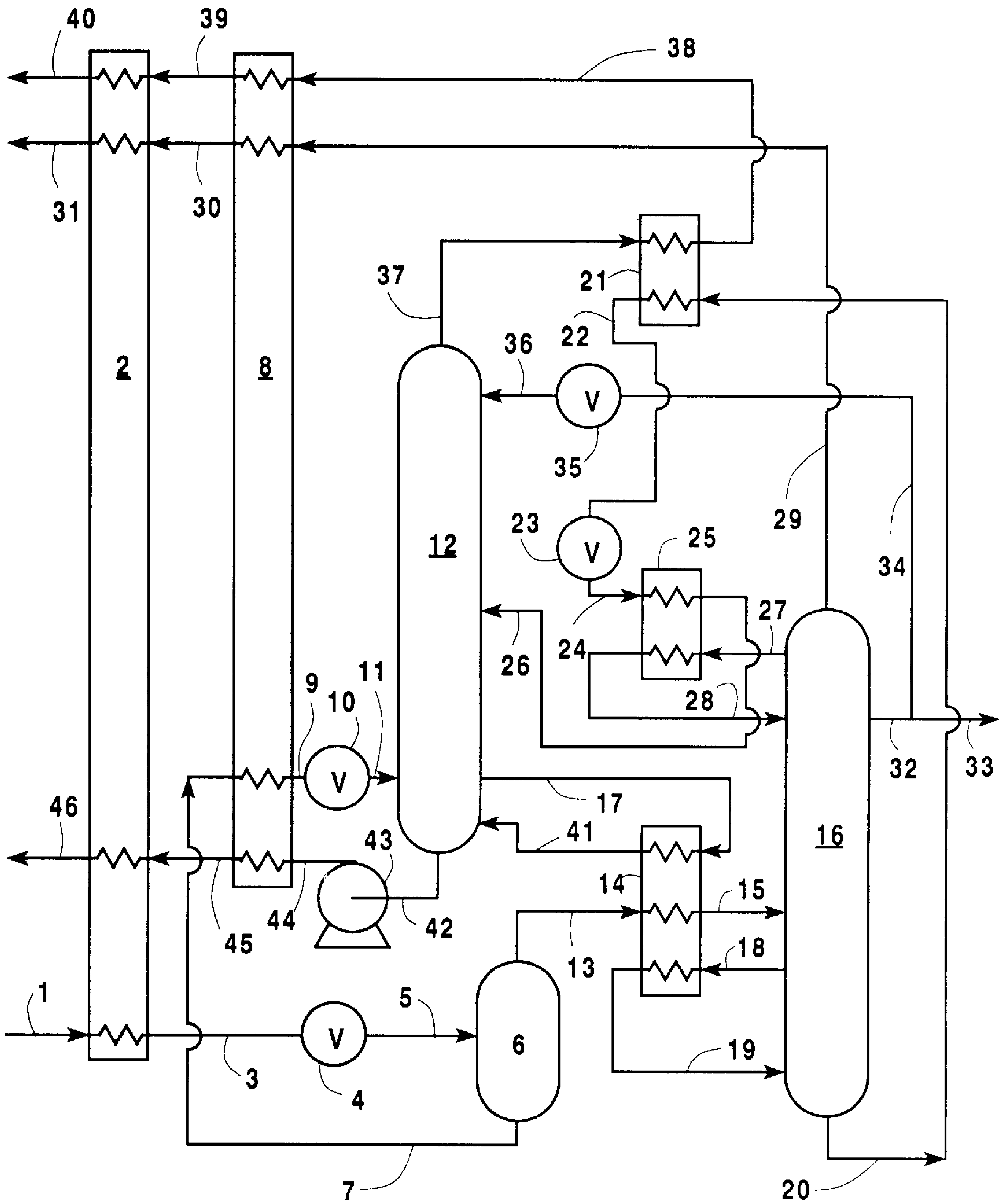
[57] **ABSTRACT**

The production of higher purity helium wherein a feed comprising helium, nitrogen and hydrocarbons is processed in a system comprising two columns operating at different pressure levels, with the product helium produced in the higher pressure column and nitrogen and hydrocarbons cycled to the lower pressure column.

7 Claims, 1 Drawing Sheet





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CRYOGENIC RECTIFICATION SYSTEM FOR PRODUCING HIGHER PURITY HELIUM

TECHNICAL FIELD

This invention relates generally to the production of helium and, more particularly, to the production of helium by cryogenic rectification.

BACKGROUND OF THE INVENTION

Heretofore, crude helium has been produced from natural gas to a purity level of about 50 percent and then passed on to a helium refinery for the production of refined helium. This relatively standard purity for crude helium is the result government requirements, and helium production facilities have historically been designed to produce helium at this purity level for passage on to a helium refinery.

The higher the purity and the higher the pressure of the helium passed on to the helium refinery, the less is the energy required to operate the helium refinery. However, typical helium production systems cannot effectively produce helium with a purity much higher than about 50 percent.

Accordingly, it is an object of this invention to provide a system for producing helium by separation from hydrocarbon fluid at a higher purity than that possible with conventional systems.

SUMMARY OF THE INVENTION

The above and other objects, which will become apparent to one skilled in the art upon a reading of this disclosure, are attained by the present invention, one aspect of which is:

A method for producing higher purity helium by cryogenic rectification comprising:

(A) processing a feed comprising helium, nitrogen and hydrocarbons to produce a first fluid enriched in hydrocarbons, and a second fluid enriched in helium;

(B) passing the first fluid into a lower pressure column and passing the second fluid into a higher pressure column;

(C) separating the second fluid within the higher pressure column by cryogenic rectification to produce a helium-richer fluid, a nitrogen-richer fluid and a hydrocarbons-richer fluid;

(D) passing nitrogen-richer fluid and hydrocarbons-richer fluid from the higher pressure column into the lower pressure column; and

(E) recovering helium-richer fluid from the higher pressure column as product higher purity helium.

Another aspect of the invention is:

Apparatus for producing higher purity helium by cryogenic rectification comprising:

(A) means for dividing a feed comprising helium, nitrogen and hydrocarbons into a first fluid enriched in hydrocarbons, and into a second fluid enriched in helium;

(B) a first column and means for passing the first fluid into the first column;

(C) a second column and means for passing the second fluid into the second column;

(D) means for passing fluid from the upper portion of the second column into the first column and means for passing fluid from the lower portion of the second column into the first column; and

(E) means for recovering fluid from the upper portion of the second column as product higher purity helium.

As used herein, the term "hydrocarbons" means one or more hydrocarbon species.

As used herein, the term "column" means a distillation or fractionation column or zone, i.e. a contacting column or zone, wherein liquid and vapor phases are countercurrently contacted to effect separation of a fluid mixture, as for example, by contacting of the vapor and liquid phases on a series of vertically spaced trays or plates mounted within the column and/or on packing elements such as structured or random packing. For a further discussion of distillation columns, see the Chemical Engineer's Handbook, fifth edition, edited by R. H. Perry and C. H. Chilton, McGraw-Hill Book Company, New York, Section **13**, *The Continuous Distillation Process.*

Vapor and liquid contacting separation processes depend on the difference in vapor pressures for the components. The high vapor pressure (or more volatile or low boiling) component will tend to concentrate in the vapor phase whereas the low vapor pressure (or less volatile or high boiling) component will tend to concentrate in the liquid phase. Partial condensation is the separation process whereby cooling of a vapor mixture can be used to concentrate the volatile component(s) in the vapor phase and thereby the less volatile component(s) in the liquid phase. Rectification, or continuous distillation, is the separation process that combines successive partial vaporizations and condensations as obtained by a countercurrent treatment of the vapor and liquid phases. The countercurrent contacting of the vapor and liquid phases is generally adiabatic and can include integral (stagewise) or differential (continuous) contact between the phases. Separation process arrangements that utilize the principles of rectification to separate mixtures are often interchangeably termed rectification columns, distillation columns, or fractionation columns. Cryogenic rectification is a rectification process carried out at least in part at temperatures at or below 160 degrees Kelvin (K).

As used herein, the term "indirect heat exchange" means the bringing of two fluid streams into heat exchange relation without any physical contact or intermixing of the fluids with each other.

As used herein, the terms "upper portion" and "lower portion" means those sections of a column respectively above and below the midpoint of the column.

As used herein, the term "tray" means a contacting stage, which is not necessarily an equilibrium stage, and may mean other contacting apparatus such as packing having a separation capability equivalent to one tray.

As used herein, the term "equilibrium stage" means a vapor-liquid contacting stage whereby the vapor and liquid leaving the stage are in mass transfer equilibrium, e.g. a tray having 100 percent efficiency or a packing element height equivalent to one theoretical plate (HETP).

BRIEF DESCRIPTION OF THE DRAWING

The sole FIGURE is a schematic representation of one preferred embodiment of the higher purity helium production system of this invention.

DETAILED DESCRIPTION

The invention enables the production of higher purity helium, typically having a purity of 70 mole percent or more, from a hydrocarbon feedstock for passage on to a helium refinery for the production of refined helium. In a particularly preferred embodiment, the invention additionally produces liquid nitrogen which may also be used in the helium refinery.

The invention will be described in greater detail with reference to the Drawing. Referring now to the Figure, feed stream **1**, which is at a pressure generally within the range of from 100 to 2000 pounds per square inch absolute (psia), is cooled by passage through heat exchanger **2** by indirect heat exchange with return streams to form cooled feed stream **3**. Feed stream **1** is a vapor comprising helium, nitrogen and hydrocarbons. Helium is present in feed stream **1** in a concentration within the range of from 0.1 to 10 mole percent; nitrogen is present in feed stream **1** in a concentration within the range of from 5 to 79.9 mole percent; hydrocarbons are present in feed stream **1** in a concentration within the range of from 20 to 94.9 mole percent. The hydrocarbons may comprise solely methane or may comprise methane along with ethane and one or more other heavier constituents commonly found in natural gas. Feed stream **1** may also contain one or more lower boiling or more volatile components such as hydrogen, neon, oxygen and argon.

Cooled feed steam **3** is passed from heat exchanger **2** to valve **4** through which it is throttled to form two phase stream **5**. Phase separator **6** receives stream **5** and within phase separator **6** the fluid in stream **5** is separated into a liquid or first fluid which is enriched in hydrocarbons, i.e. having a hydrocarbons concentration which exceeds that of feed stream **1**, and into a vapor or second fluid which is enriched in helium, i.e. having a helium concentration which exceeds that of feed stream **1**. The first fluid, which also contains nitrogen and may contain trace amounts of helium, is passed from phase separator **6** as stream **7** through heat exchanger **8** wherein it is subcooled by indirect heat exchange with return streams. Resulting subcooled stream **9** is flashed across valve **10** and then passed as stream **11** into first or lower pressure column which is operating at a pressure generally within the range of from 15 to 200 psia.

The second fluid, which also contains nitrogen and hydrocarbons, is passed from phase separator **6** as stream **13** through heat exchanger **14** wherein it is partially condensed. Resulting partially condensed stream **15** is then passed as feed into second or higher pressure column **16** which is operating at a pressure higher than that of lower pressure column **12** and generally within the range of from 100 to 500 psia.

Within higher pressure column **16** the feed is separated by cryogenic rectification into a helium-richer fluid, a nitrogen-richer fluid and a hydrocarbons-richer fluid. A liquid stream **18** is withdrawn from column **16** and partially vaporized by passage through heat exchanger **14** by indirect heat exchange with the aforesaid partially condensing second fluid. Resulting two-phase stream **19** is returned to column **16**. The vapor portion of stream **19** provides vapor upflow for column **16**. The liquid portion of stream **19** comprises hydrocarbons-richer fluid. Hydrocarbons-richer fluid is withdrawn from the lower portion of column **16** as stream **20** and subcooled by passage through heat exchanger **21**. Resulting subcooled stream **22** is throttled across valve **23** and then passed as stream **24** through heat exchanger **25** wherein it is at least partially vaporized. Resulting fluid is passed in stream **26** from heat exchange **25** into lower pressure column **12**.

The fluid comprising stream **24** is at least partially vaporized in heat exchanger **25** by indirect heat exchange with a vapor passed in stream **27** from column **16** through heat exchanger **25** wherein it is partially condensed. Resulting two-phase stream **28** is returned to column **16**. The liquid portion of stream **28** provides liquid downflow for column **16**. The vapor portion of stream **28** comprises helium-richer fluid. Helium-richer fluid is withdrawn from the upper portion of column **16** as stream **29** and warmed by passage through heat exchanger **8**. Resulting warmed helium-richer stream **30** is further warmed by passage through heat exchanger **2** and recovered as product higher purity helium **31** having a helium concentration of at least **65** mole percent and generally having a helium concentration within the range of from 70 to 95 mole percent. Preferably higher purity helium **31** is recovered at an elevated pressure, generally within the range of from 100 to 400 psia. Higher purity helium **31** may be passed on to a helium refinery as a feed stream for the production of refined helium.

Nitrogen-richer fluid is withdrawn as liquid in stream **32** from the upper portion of higher pressure column **16** at a level generally not more than ten equilibrium stages below the level from which helium-richer fluid in stream **29** is withdrawn from column **16**. Preferably the nitrogen-richer fluid is withdrawn from column **16** at the same equilibrium stage as that from which helium-richer stream **29** is withdrawn. The nitrogen-richer fluid will generally have a nitrogen concentration of at least 95 mole percent. Preferably, as illustrated in the Figure, a portion **33** of stream **32** is recovered as product liquid nitrogen. This product liquid nitrogen may conveniently be employed as refrigerant in a helium liquefier. The remaining portion **34** of stream **32** may be throttled across valve **35** and passed as stream **36** into lower pressure column **12** as reflux.

Within column **12** the feeds are separated by cryogenic rectification into nitrogen vapor and hydrocarbon liquid. The nitrogen vapor is withdrawn from column **12** as stream **37** and warmed by passage through heat exchanger **21** by indirect heat exchange with the subcooling hydrocarbons-richer fluid in stream **20**. Resulting warmed nitrogen vapor **38** is warmed by passage through heat exchanger **8**, resulting stream **39** is further warmed by passage through heat exchanger **2**, and resulting nitrogen vapor stream **40** is passed out of the system. Stream **40** may be released to the atmosphere or may be recovered, in whole or in part, such as for use such as in secondary hydrocarbon recovery.

A liquid stream is removed from column **12** as stream **17** and passed through heat exchanger **14** wherein it is partially vaporized by indirect heat exchange with the aforesaid partially condensing second fluid **13**. Resulting two phase stream **41** is returned to column **12**. The vapor portion of stream **41** provides vapor upflow for column **12**. The liquid portion of stream **41** comprises hydrocarbon liquid. The hydrocarbon liquid is withdrawn from the lower portion of column **12** as stream **42**. Preferably the hydrocarbon liquid is increased in pressure, such as by passage through liquid pump **43** as illustrated in the Figure. Resulting pressurized hydrocarbon liquid **44** is warmed by passage through heat exchanger **8** and resulting stream **45** is warmed and preferably vaporized by passage through heat exchanger **2**. Resulting hydrocarbon stream **46** is recovered as product methane or natural gas having a hydrocarbons concentration generally within the range of from 90 to 100 mole percent.

Table 1 presents the results of a computer simulation of one example of the invention, using an embodiment similar to that illustrated in the Figure. The stream numbers in Table 1 correspond to those of the Figure. This example is presented for illustrative purposes and is not intended to be limiting.

TABLE 1

Stream No.	Flowrate (lbmol/hr)	Temperature K	Pressure (psia)	composition (mole %) He	N2	C1 plus
1	1000	160	400	2.0	58.0	40.0
46	414	150	100	0.0	4.0	96.0
31	26	150	400	75.0	25.0	0.0
40	560	150	30	0.1	99.4	0.5

As can be seen from the example reported in Table 1, the invention enables the production of helium separated from a hydrocarbon fluid, having a concentration significantly higher than that attainable effectively with conventional processes. In the specific example reported in Table 1, the product higher purity helium has a helium concentration of 75 mole percent from a feed stream having a helium concentration of only 2 mole percent.

Although the invention has been described in detail with reference to a certain preferred embodiment, those skilled in the art will recognize that there are other embodiments of the invention within the spirit and the scope of the claims.

We claim:

1. A method for producing higher purity helium by cryogenic rectification comprising:

(A) processing a feed comprising helium, nitrogen and hydrocarbons to produce a first fluid enriched in hydrocarbons, and a second fluid enriched in helium;

(B) passing the first fluid into a lower pressure column and passing the second fluid into a higher pressure column;

(C) separating the second fluid within the higher pressure column by cryogenic rectification to produce a helium-richer fluid, a nitrogen-richer fluid and a hydrocarbons-richer fluid;

(D) passing nitrogen-richer fluid and hydrocarbons-richer fluid from the higher pressure column into the lower pressure column; and

(E) recovering helium-richer fluid from the higher pressure column as product higher purity helium.

2. The method of claim **1** wherein the product higher purity helium has a helium concentration of at least 65 mole percent.

3. The method of claim **1** further comprising recovering a portion of the nitrogen-richer fluid from the higher pressure column.

4. The method of claim **1** further comprising producing nitrogen vapor within the lower pressure column and recovering nitrogen vapor from the upper portion of the lower pressure column.

5. The method of claim **1** further comprising producing hydrocarbon liquid within the lower pressure column, withdrawing hydrocarbon liquid from the lower portion of the lower pressure column, and recovering resulting fluid as product having a hydrocarbons concentration within the range of from 90 to 100 mole percent.

6. Apparatus for producing higher purity helium by cryogenic rectification comprising:

(A) means for dividing a feed comprising helium, nitrogen and hydrocarbons into a first fluid enriched in hydrocarbons, and into a second fluid enriched in helium;

(B) a first column and means for passing the first fluid into the first column;

(C) a second column and means for passing the second fluid into the second column;

(D) means for passing fluid from the upper portion of the second column into the first column and means for passing fluid from the lower portion of the second column into the first column; and

(E) means for recovering fluid from the upper portion of the second column as product higher purity helium.

7. The apparatus of claim **6** further comprising means for recovering fluid from the lower portion of the first column.

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